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PROGRESS REPORT ON THE DETERMINATION OF ALPHA EMITTERS IN AIR

by

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INTRODUCTION

An electrostatic precipitator and a newly developed filter paper are compared for efficiency, reliability, availability, and ease of use. The aerosols collected in either case are counted for alpha activity in a cylindrical pulse chamber, the design of which is an adaptation of the breech-locking air alpha chamber. Vapor and gases are not collected by either instrument, unless occluded on particulate matter.

I. THE ELECTROSTATIC PRECIPITATOR

The electrostatic precipitator has become one of the standard instruments used to collect dust from air.[1] The Mine Safety Appliance Company manufactures a small, portable "Precipitron" (using preionization before collection) which was designed originally by Westinghouse for the study of dust hazards. The instrument was not found dependable for continuous operation and procurement was very slow, so another design was built at Clinton by the Instrument Shop. The collecting electrode arrangement used by MSA was duplicated, but the electrical circuit and fan were changed. These instruments are improvements, and have been used continuously for several months, but further improvement would be desirable.

Recently, the Westinghouse Research Laboratory and the Bureau of Mines Research Laboratory were visited to obtain first-hand information about the precipitron and dust sampling techniques. From these visits and experiences with the precipitron at Clinton the following conclusions can be drawn.

a) The precipitrons built at Clinton, while an improvement over MSA model, require frequent repairs. The vibrators, coils, and fans have to be replaced frequently, and the weight and size are considered too large. No light-weight high-voltage supply has been found commercially available, and a more satisfactory fan has not been procured. Several possible sources of high voltage have been suggested, but not investigated. These are an oil furnace ignition coil, a small neon sight transformer with the secondary center not tapped to ground, and an rf oscillator and air-core transformer such as RCA uses in television work.

b) Particles which are collected on the collecting electrode can be recharged and repelled into the air stream again. For certain kinds of particles, this may become a serious problem, because the particles tend to repeatedly reverse their charge, and pass through the collector. The Westinghouse people believe that lining the collecting cylinder with paper might tend to increase the reverse ionization and interfere with the action of the precipitron in other ways. Paper liners are used almost all the time at Clinton and no great difficulty has occurred. Unexplained failures of the precipitrons have been observed, however, but even when aluminum foil liners were used. An adhesive coating on the liner might help if one could be found that did not interfere with the alpha counting.

c) For high efficiences, the velocity of the air through the collecting system should not exceed 150 linear feet per minute. This is equivalent to 1.8 cubic feet per minute through the $1\frac{1}{2}$ inch diameter collector now in use. The rate used at Clinton has been usually about 11 cubic feet per minute. Parker has determined the efficiency at this rate to be about 70%.[2] Westinghouse believes that the dimensions of the collecting system cannot be enlarged, as they are the result of a compromise between factors of field strength, sparking distance, and practical experience.

d) No data has been published or collected, as far as can be determined, on the collection of small particles. All accurate data is limited to the sizes visible in a microscope, and even then the question of real vs apparent size is involved at the lower limits. The Bureau of Mines tend to be skeptical about sizes below 0.5 micron. The technique of measuring aerosol particles with the electron microscope is just being worked out. It is hoped that more definite information can be obtained at Chicago using these techniques.

e) The toxicity of radioactive material is higher than that of materials for which the MSA precipitron was designed.

f) High voltage discharges in the precipitron sometimes interfere with nearby counters.

II. THE COUNTING CHAMBER

Since the precipitator collects on the inside surface of a cylinder, an alpha counting chamber was constructed which consists of a slightly larger cylinder as the high voltage electrode, and a concentric rod which acts as the pulse collecting electrode. Except for the extra length due to the cylindrical electrodes, the appearance, the amplifier mounting, and the operation are the same as that of the standard alpha counting chamber. Microphonics and arcing due to fuzz and hairs are reduced to a satisfactory level in this chamber. Complete constructional details are available.

III. FILTER PAPER COLLECTION

Special high efficiency air filtering papers procured through NDRC have been found practical and greatly superior to ordinary papers such as Whatman 50, which was previously tested. The paper is a mixture of ordinary cellulose fibers (about 20 microns diameter) and specially prepared asbestos fibers with diameters of one micron and less, which mat between the cellulose fibers and do the actual collecting. The efficiency and resistance of this type paper depends on the ratio of asbestos to cellulose, as well as the thickness.

The thinnest and smoothest paper of this type (made for the Navy) was selected for experimentation in the present monitoring problem. This paper passes less than 0.1% of a smoke with average particle size of approximately 0.4 microns, according to the scientists of the NDRC Division 10, who have developed and tested it extensively.

There is naturally a question of alpha absorption which depends on how deeply the collected particles are buried in the paper. For this reason, the thinnest, hardest packed papers which still have a satisfactory resistance and efficiency, are desirable. The Navy paper presently in use was tested for absorption losses. From this data we can conclude that the loss is unlikely to be more than 1/3 of the actual count and maybe less. This is judged to be satisfactory for this type of work; a loss of 2/3 or so would have been open to question. A special paper is being ordered for our work which will probably have much lower absorption, equally high efficiency, but greater resistance.

One interesting test was made with an experimental paper consisting of 100% of the fine asbestos fibers, floated out on light tissue as a support. This paper is impractical for routine use because of its lack of tenacious qualities, and the difficulties in producing it. Several sheets were placed in series and air containing suspended product particles was pulled through. (This is one of the standard methods of measuring the efficiency.) Upon counting, it was found that all the activity remained on the first sheet.

Furthermore, it was located very close to the surface of this sheet. The reverse side of the sheet counted about half as high as the top side, and when another piece of the same paper was put on top of the contaminated paper, the count was again reduced by about one half. The paper chosen for routine use, then will have as high as possible percentage of the asbestos fibers, and be as thin as practical.

A holder has been designed which directs the air to be tested into the inside surface of a cylinder of filter paper. This paper can then be slipped into the cylindrical alpha counter and "counted" in the same manner as the precipitron-collected samples. (See photograph.) Also, a quiet-running vacuum cleaner blower has been found which easily has the capacity necessary to pull three or four cubic feet per minute through the new tester against a resistance of several times that introduced by the Navy paper samples presently in use. These blowers are being ordered, and will be fitted with supports to hold the cylindrical filter paper holder and air flow indicator, thus forming a very portable and easy to use air testing unit. A new filter paper is being ordered which will be only about 0.010-inch thick and will have a high percentage of asbestos fibers. This should have high collection efficiency and low absorption loss. Experiments to establish the absorption factor will be performed when the paper is received.

IV. COMPARISON TESTS

The electrostatic precipitator was tested in series and in parallel with the Navy paper, under large scale process and ordinary laboratory conditions, as well as under controlled conditions using a dry box and artificially prepared product suspensions. Some tests were also performed using the alpha emitters which occur naturally in the air.

The results of these tests indicate that at no time did any significant quantity of active material pass through the paper. In most cases, some material apparently did pass through the precipitator. In parallel tests drawing air through the instruments at the same rate, the counts were nearly the same, indicating that the absorption loss of the paper and inefficiency of the precipitron may be about equal. The choice then becomes one of reliability and convenience. For reasons already stated a better precipitron should be designed if this is to be used.

V. GENERAL INFORMATION

To make and seal the filter paper cylinders, a special loading fixture has been designed. This consists of a brass tube supported at one end, with spring brass clamps to hold the paper sheet in the form of a cylinder while cellulose tape is applied to form the seal down its length, on both the inside and the outside. The photograph shows this tool as well as the new filter paper holder.

A complete apparatus for sampling the air for alpha emitters would consist of several filter paper holders, extra support screens, a blower for each holder, connecting hose and clamps for convenience, a loader, and a special cylindrical alpha chamber with the standard amplifier and scaler.

A suggested way to conduct air surveys is to have the samplers portable, and with them locate the actual sources of alpha contamination by frequent "spot checks" in the various laboratories. Particular apparatus such as centrifuges, and fuming and transfer processes should be carefully monitored. Permanent installations of samplers in rooms or cells containing especially questionable processes would be advisable, with tests run at intervals every day, or during the suspected operations. Also, samplers might be placed to test the exit air of the ventilation ducts in an attempt to find which particular rooms release the most contamination.

It is possible for an instrument to be designed which would integrate the activity as it is collected. Probably such an instrument would have to use a proportional counter and rate meter to be as sensitive and dependable as desired. The original "Sneezy" employed a DC ion chamber and amplifier, but it proved impractical. The difficulty is that radium and thorium disintegration products normally present in the air collected also. The amount of short-lived activity from these sources is often much greater

than the amount of product activity believed to be tolerable in air. This natural activity is subject to wide variations. Therefore, it would have to be corrected for, simultaneously with collection. The alternative would be to allow this activity to decay before counting. The average half-life of the radium products is about 40 minutes, and the thorium about 10.6 hours.[4] Several instruments have been proposed which would correct for the natural activity in one of these two ways, but no models have been built. If an instrument which will alarm when gross amounts of activity are present in the sampled air is desired, possibly the alarming level would be such that the natural activity could be ignored. Appendix III indicates the order of magnitude of the natural activity which can be expected.

Usually activity found in the air is quite localized, and can be traced to a particular place or operation. This is especially true in laboratories where there are several hoods and the air is changing at a high rate. In large operations or badly contaminated rooms, it is possible to observe a rise in the general contamination of the air. This is usually of lower magnitude than that found locally, but large enough to be significant. A permanently installed integrating monitor may be of value in such a room, but still no assurance can be given as to protection from localized concentrations which the device may easily miss.

It much be emphasized that both the precipitron and filter paper collect only particulate matter, down to colloidal sizes, but vapors or gases are usually not collected. Cases such as radon may be exceptions, if they are adsorbed on particles of dust.

For their help in this development, acknowledgement is given to William Morris and George Koval at Clinton and Albert Potts and Ralph Gardiner at Chicago.

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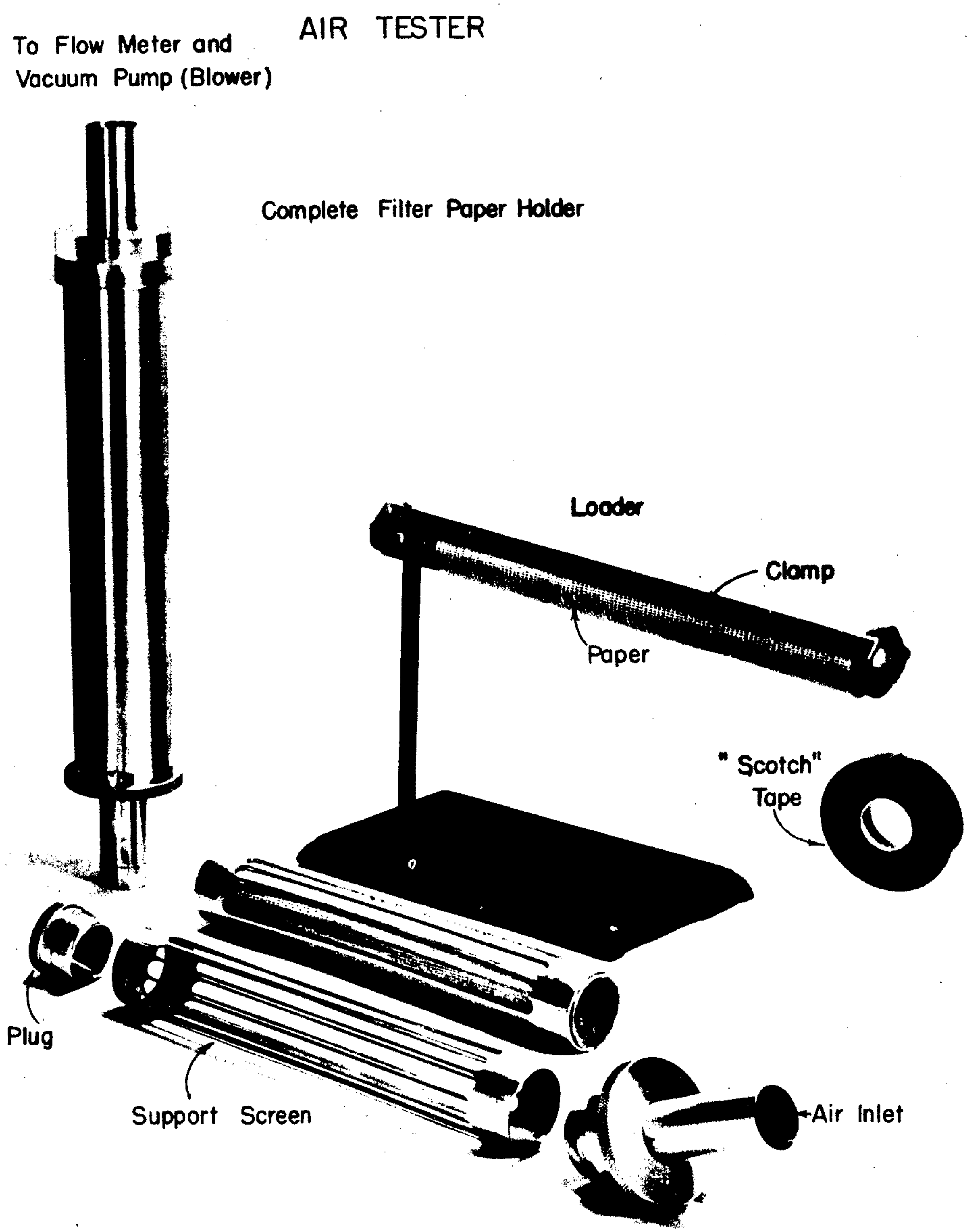

Figure 1. Air tester.

APPENDIX

I. ABSORPTION TESTS DATA

In general, the procedure was to pull contaminated air through the filter paper, take a direct alpha count of the paper, then chemically digest and extract the active material from the paper on to a platinum plate, and compare the counts.

Collection

Contaminated air was created by bubbling air through a solution containing several grams of product. Air was pulled through filter discs 4.25 cm diameter at about 6 liters/minute which gives a linear velocity at the paper of about 200 feet/minute, which in turn is about the rate resulting from pulling 3 cubic feet/minute through the larger cylindrical tester.

The collection periods varied from 20 to 40 minutes. The rate of air bubbling through solution was increased when not much activity was collected at the first two trials.

Extraction

Two methods were employed; one a precipitation and the other a simple evaporation.

The precipitation process was tested by applying a known amount of activity in tracer quantities, whereby about 85% recovery was obtained. Blanks (similar but clean papers) were run parallel with the significant papers to test for contamination from reagents or other causes. In every case the blanks yielded less than 5 counts/minute, with chamber background subtracted. In all cases the 85% recovery and the background were applied as corrections.

In the evaporation method, the total supernatant volume was reduced to about 3 ml and then transferred to a larger platinum plate for the final evaporation.

Results: Standard high-geometry chamber used for all counts. Counting periods 8 minutes for low counts.

Test No.	Direct count before extraction counts/minutes	Method	Corrected count after extraction	% Difference
102	12	Evaporation	17	+30%
103	32	Precipitation	36	+11
104	3	"	5	--
105	13	Evaporation	18	+28
106	2730	Precipitation	5060	+46%
107	870	Evaporation	875	+ 0.6%
108	250	Evaporation	208	-20
109	175	Precipitation	230	+24
110	150	"	201	+25
111	190	Evaporation	198	+ 4

II. COMPARISON TESTS

A. Using Natural Alpha Activity

Although the particle sizes of the alpha emitting dusts in the normal air is not known, the collection of this fast-decaying activity was a simple means for comparing the precipitator and the paper collection methods. The radon and thoron gases and disintegration products are perhaps adsorbed on the fairly

large particles of carbon soot, silicon, and whatever else constitutes "city dust." Precipitators and paper collectors were set up in series and in parallel for these tests, and the amount of accumulated material judged from extrapolations of the decay curves to 0 time, the end of the collection period in each case.

The following is a summary of the results of these tests.

Series Tests

Non-airconditioned room (Chicago)
3 hours at 2 cubic feet/minute.
0 time is end of collection period.

Key:

Test A. Filter paper (Navy) first, then precipitator
A-1 Activity on filter paper
A-2 Activity on precipitator

Test B. Precipitator first, then filter paper. (Done on a different day, so total collected is not significant)
B-1 Activity on precipitator
B-2 Activity on filter paper

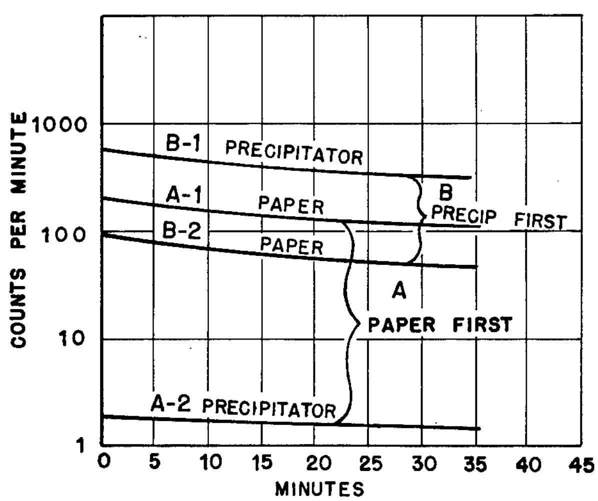

Figure 2.

This test was repeated several times with similar results except for one case in which five times as much activity was found on the paper even though it followed the precipitator. The assumption is that while the apparatus was checked occasionally and seemed to be operating normally (visable corona discharge), it must have stopped occasionally or become inefficient because of too dry air or some unknown cause.

The following graph represents a test wherein a precipitator and the paper were run in series in that order, while another precipitator ran in parallel; also the reverse case of the paper first and then the precipitator in series, with the other precipitator in parallel.

All the runs were for 110 minutes.

0 time is at the end of the collection period.

All runs made at the same time and place. (Clinton, hallway of Chemistry Building, run B immediately following run A.)

Key:

Test A - Filter paper first, then precipitator

A-1 - Filter paper, at 1.5 cfm.

A-2 - Precipitator following A-1

A-3 - Precipitator at 11.5 cfm.

Test B

B-1 - Precipitator, at 1.5 cfm.

B-2 - Filter paper following B-1.

B-3 - Precipitator, at 11.5 cfm.

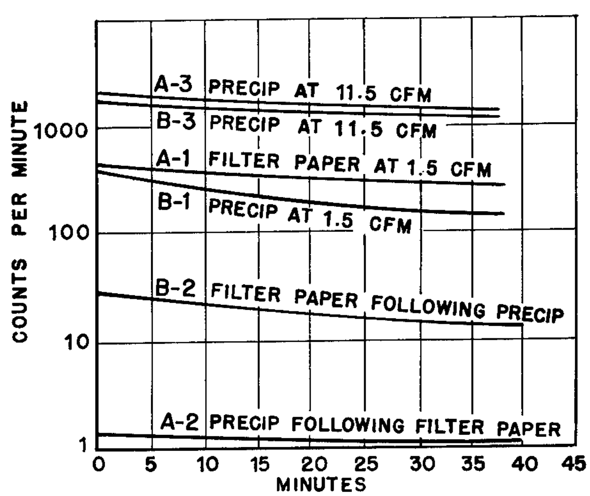

Figure 3.

B. Using Contaminated Air

Two precipitators and a paper holder were also used in the experiments described in Appendix I.

The counts given here are those taken six days after the tests were made so the natural background has disappeared. They were made on the special cylindrical alpha counter described above.

Test No.	Type of Test	Flow rate cubic ft per minute	Time of run	Alpha counts per minute
200	Navy Paper	1.06	20 min	81
A 300	Precipitator	1.06	20	56
301	Precipitator	11	20	1054
201	Navy Paper	0.94	40	38
B 302	Precipitator	0.94	40	32
303	Precipitator	11	40	544
202	Navy Paper	1.09	35	4880
C 304	Precipitator	1.09	35	6024
305	Precipitator	11	35	10,500 (?)
203	Navy Paper	0.68	30	206
D 306	Precipitator	0.68	30	120
307	Precipitator	11	30	5500

The test number 305 was so "hot" that the count is not reliable. When this paper was cut into small pieces for separate counting, some of these pieces nearly jammed the counter. It is interesting to note that the pieces cut out of the back edge of the deposition tube were nearly as active as any other piece, indicating that much material must have passed on through. Little activity would be expected at the trailing end of the cylinder if high efficiencies were the case.

Since the filter paper has not been standardized, the different pieces used in this series of tests had different resistances. The blower fan employed was simply the fan of one of the precipitators (with the high voltage off), so the flow rates obtained were considerably lower than that expected with the blowers now being procured. One of the precipitators running parallel to the filter paper in each case was adjusted for the same flow as that obtained through the paper, and the other run at full speed, as has been the custom. Calibrated orfices were used for flow-meters.

The uncertainty of the degree of air contamination will perhaps account for the fact that the precipitron running at 11 cfm collected more than 11 times the activity than the precipitron and paper running at about 1 cfm. It is likely that at the higher rate a greater quantity of the active solution droplets which normally would not rise out of the tank were drawn up to the collector by the increased draft.

C. Using Dry Box Controlled Conditions

A metal box of cubic shape, 20 inches on each side with a glass roof, attached rubber gloves for inside manipulations, air-tight door, and air inlet and outlet, was set up for testing various papers and comparing means of collecting. Several experiments were performed, the results of which are given here; but the main function of this work was to develop a satisfactory means of producing product dusts of extremely small particle size (from 0.1 to 1 micron) to be used in animal inhalation experiments.(Work of Dr. Albert Potts at Chicago.) This first work was not very successful, but it led to a method which at present seems quite efficient and will be used in future experiments, especially with the special "tailor made" paper when it arrives.

A "bomb" consisting of a strong seamless tube about 8 inches long fitted with a needle valve and nozzle at one end and a plug at the other is placed in a freezing solution. Gaseous Freon is then condensed into the "bomb" after which a chloroform extraction of a product cupferide ion is introduced. Upon reaching room temperature, a tremendous pressure had developed, so when the valve was opened (after the air flow through the box was established and the filters to be tested placed on the outlet), a spray could be observed from which the product was left suspended in presumably minute particles.

A definite Tyndall effect was observed each time. Samples were taken with a thermal precipitator which were later observed with an electron microscope. It was thus found that the particle size ranged from 0 to 2 microns, with a predominance of the larger sizes. Subsequent work with other types of Freon and other chemical methods of preparing the soluble product ions have considerably improved upon the original low efficiency, and can produce more uniform small particles. More conclusive penetration tests are now possible, and will be performed. Special atomizers are also being tried.

The following is a summary of the results of the tests with the box:

For easier control, and to insure a dust-free atmosphere before releasing the "bomb," N_2 gas was used to flush the box and also to flow through the inlet. Flow rate through box (with filter papers on the outlet) was about 4.5 liters/minute, using 4.25 cm filter discs.

The length of time of the tests is unimportant since the Tyndall effect became nil in about 10 minutes after the test started, and the tests were all for at least 20 minutes. For this timing about 90 liters of air were tested out of the 130 liters contained in the box. Concentrations of product dust were both unknown and constantly changing, so only the relative activities of the successive papers should be considered.

Test 1.

4 sheets of Whatman 50 in series

Top sheet No. 1 - 575 counts/minute
2 - 38
3 - 23
4 - 12

Test 2.

4 sheets of Navy asbestos type filter paper in series

Top sheet No. 1 - 1440 c/m
2 - 3
3 - 2 (Background)
4 - 3

Test 3.

4 sheets special thin 100% asbestos fiber paper in series

Top sheet No. 1 - 1520 c/m
2 - 5
3 - 2 (Background)
4 - 2

	1st Count	2nd Count
Sheet No. 1 - off top -	1520	1550
Reversed -	768	736
"Hot" side up but covered with similar sheet-	771	733

This shows all the activity was near the top surface.

Test 4.

Whatman 50	- 84 c/m	In series - 4 min test.
Navy paper	5 c/m	
Navy paper	- 64 c/m	In series - 4 min.
Whatman 50	1 c/m	(Concentration in box now less)
100% asbestos paper	- 572 c/m	
same	1	In series 20 minutes
same	1	

Test 5.

Navy paper	- 5,240 c/m	
El. Precipitator	3 c/m	In series - larger paper used, flow at 2 cfm.

Test 6.

El. Precipitator	- 7,800 c/m	In series.
Navy paper		

These tests were performed mainly to perfect an effective means of producing a fine product suspension, but the data indicates the effectiveness of the new asbestos Navy paper.

III. NATURAL ALPHA BACKGROUND

Considerable work has been done by Parker and others in identifying the natural materials which give rise to the short-lived alpha activity collected in every case. The general conclusions are that it is the result of the disintegration products of radon and thoron, gasses which are continually supplied to the atmosphere from the rocks. Generally, there is from 20 to 100 times as much radon as thoron. The significant disintegration chains are:

(Fast) $\mathrm{Rn} \xrightarrow[3.8d]{\alpha} \mathrm{Ra\ A} \xrightarrow[3.05m]{\alpha} \mathrm{Ra\ B} \xrightarrow[26.8m]{\beta} \mathrm{Ra\ C} <$ (further disintegrations unimportant for this work)

(Slow) $\mathrm{Th} \xrightarrow[54.5s]{\alpha} \mathrm{Th\ A} \xrightarrow[0.518s]{\alpha} \mathrm{Th\ B} \xrightarrow[10.6h]{\beta} \mathrm{Th\ C} <$

Since a decaying activity is reduced to about 1% of its original value in less than 7 half-lives, we can disregard the radium series if we wait about three hours before making a measurement. The thorium series requires three or four days to decay to an insignificant level, but a formula may be used to discount this activity from any two readings, thus obtaining a value for the activity due to any long-lived alpha emitters collected. The first of these two readings must be made after about four hours has elapsed, for that is the time required for the equilibrium condition of Th B to be achieved and the decay to start. This is especially true for a short collection period, when a rise in activity could be observed during the several hours immediately after the collection period, if the decaying activity due to the radium series were not so great.

This formula is:

Long-lived activity in counts/minute

$$\frac{C_2 - C_1 e^{-\lambda \Delta t}}{1 - e^{-\lambda \Delta t}}$$

C_1 = first count (c/m) made after 4 hours

C_2 = second count

Δt = hours between counts (usually about 24)

λ = 0.06

See Chapter VIII of Hevesy and Paneth, A Manual of Radioactivity, for a discussion of the mathematics of this type of collection and decay problem.

To get an idea of how this background varies from time to time and place to place, a number of the routine tests with the electrostatic precipitator were counted within a short time after the end of the collection period. The possible instability of the precipitator could conceivably give rise to some of the variation, however. These data are summarized in the following table.

From data with precipitator:

The tests where the alpha activity eventually decayed below 20 counts/minute are cited without comment; those that retained activity due to product are specially noted.

Each test was for 30 minutes at the full $11\frac{1}{2}$ cfm.

These rooms were not air conditioned.

The time from the end of the collection period to the first count is important, but could not be maintained the same for each test; however, an indication was obtained of how the natural activity levels vary. (Some variation possibly due to precipitator characteristics.)

Date	Room at Clinton	Time end of test	Minutes until first count	Activity at 1st count c/m
10-17-44	22	11:05	15	547
,,	54	1:35	10	330
,,	54	1:38	27	26
,,	SW	12:20	35	169
,,	SW	2:55	5	297
10-19-44	54	10:25	20	795
,,	54	10:30	30	1232
,,	33-35	1:25	65	484
,,	33-35	1:35	40	692
,,	22	2:20	25	558
,,	34	2:25	25	782
,,	SW	2:45	10	4560 decayed to 3430
10-20-44	27	10:20	25	89
,,	SW	3:00	5	684 decayed to 148
,,	SW	3:40	10	77
10-21-44	SW	2:00	152	516
10-23-44	54	10:25	15	1218
,,	54	10:30	20	1406
,,		9:40	18	100
,,	SW	1:25	35	304
,,	SW	1:30	35	166
,,	SW	2:00	10	308

Date	Room at Clinton	Time end of test	Minutes until first count	Activity at 1st count c/m
10-24-44	54	10:42	18	10
"	34	10:50	15	1574
"	54	1:00	10	554
"	54	1:07	13	5
"	SW	2:10	80	184
"	22	2:45	75	12

From data with filter paper:

Tests run at Chicago. New cylindrical filter paper holder and counter used. Each test was for 30 minutes at 3 cfm.

date	room conditions	time, end of test	minutes until first count	activity at first count - c/m
12-29-44	air-conditioned laboratory	10:25	3	340
1-2-45	metal foundry	1:25	2	215
1-2-45	air-conditioned laboratory	12:15	4	228
1-2-45	office, not air-conditioned	2:35	3	220

This work is being continued on a daily basis in order better to establish the variation of the natural alpha activity as collected by the new type filter paper. A further report will be issued when enough data has been accumulated.

END OF DOCUMENT